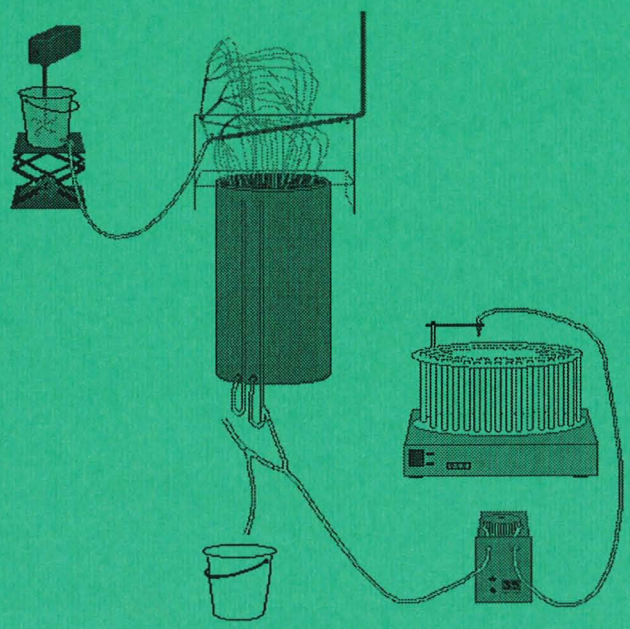
Retention of Colloids in Lysimeter Experiments on Undisturbed Macroporous Clay Soil

Linda Bengtson



Examensarbete
Handledare: Nicholas Jarvis

Institutionen för markvetenskap
Avdelningen för lantbrukets hydroteknik

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Avdelningsmeddelande 99:4
Communications

Uppsala 1999
ISSN 0282-6569
ISRN SLU-HY-AVDM--99/4--SE

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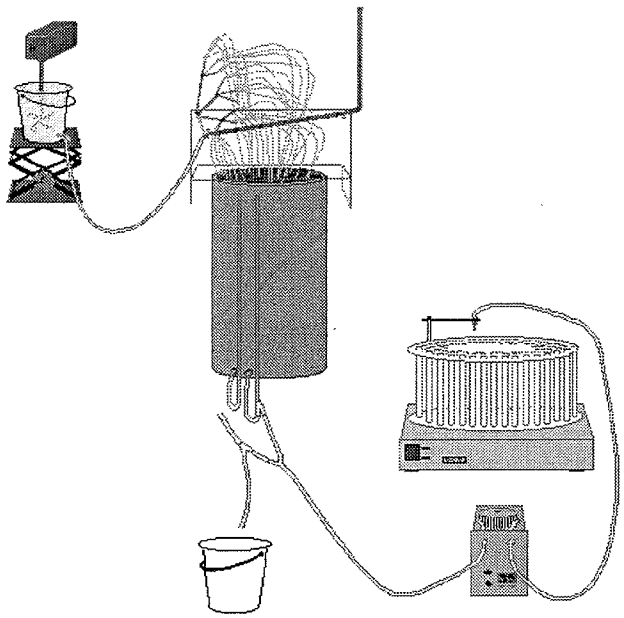

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1. ABSTRACT

Colloid-facilitated transport can cause a considerable increase in the mobility of low-solubility contaminants in soil. Preferential macropore flow in structured clay soils decreases the filtering of colloids during transport to groundwater. This experimental study was carried out to improve knowledge on how filtering is influenced by flow velocity of a percolating colloid suspension. A colloid suspension was applied with different irrigation intensities to each of four undisturbed clay soil lysimeters. Break-through curves were constructed based upon turbidity measurements on the samples, since no satisfactory calibration function between turbidity and colloid concentration could be obtained. The breakthrough curves did not reach a plateau level during the experiment. Instead, turbidity increased steadily until the application was stopped. Too low ionic strength of the water used for saturation of the lysimeters before the start of the experiments may have initiated a dispersive process causing internal colloid mobilization. Another explanation is that the relationship between turbidity and colloid concentration may depend on the particle size distribution in the effluent, which may have varied during the experiment. There is a need to further develop the methodology for working with colloid suspensions.

2. REFERAT

Kolloidburen transport av svårlösliga föroreningar orsakar en avsevärd ökning av deras rörlighet i mark. Dessutom minskas markens filtrering av kolloider i strukturerade lerjordar av så kallat 'preferential macropore flow', det makroporflöde som resulterar av att horisontella konduktiviteten är så låg att spridning i sidled av vattnet inte hinner ske vid perkolation i stora porer. Av dessa anledningar ökar risken för utlakning av bland annat fosfor, svårlösliga pesticider och tungmetaller till grundvattnet avsevärt. Detta examensarbete utfördes för att öka kunskapen om hur markfiltrering påverkas av flödeshastigheten hos en perkolerande kolloidsuspension. En kolloidsuspension påfördes med olika flödesintensiteter till var och en av fyra lysimetrar med opåverkad strukturerad lerjord. Genombrottskurvor ritades upp utifrån grumlighetsmätningar (turbiditet) på uppsamlade vattenprov från lysimeterutloppet. En tillförlitlig kalibreringskurva mellan grumlighet och kolloidkoncentration kunde inte uppnås på grund av avsaknaden av utvecklad arbetsmetod för pipettering av kolloidsuspensioner. Genombrottskurvorna uppvisade ingen tydlig platå, utan grumligheten i utflödesproverna ökade ända tills appliceringen av kolloidsuspensionen stängdes av. En förklaring kan vara för låg jonstyrka i vattnet som användes för att mätta lysimetrarna innan experimenten startade. Det kan ha haft en aggregatförsvagande verkan och startat en inre mobilisering av kolloider i lysimetern. En annan förklaring är att grumlighet representerar kolloidsammansättningen i utflödesssuspensionen - i fråga om kornstorleksfördelningen - på ett annat sätt än vad koncentration gör, eftersom små partiklar troligen ger mer utslag i grumlighet än i viktbestämning jämfört med större partiklar. Möjligheten finns också att en platå hade uppnåtts om försöket fått pågå längre. Filtreringsgraden var enligt beräkningar lägre vid den högre regnintensiteten, vilket förklaras med att större makroporer var aktiva vid perkolationen, varvid proportionellt sett färre kolloider fastnade. Det behövs ytterligare metodutveckling kring arbete med kolloidsuspensioner.

3. INTRODUCTION

Soil as a natural resource is well-known for its filtering capacity of polluted water. Contaminants dissolved in the infiltrating water or carried by particles can either be trapped in the soil body by adsorption or complex bonding, or be decomposed by soil micro-organisms. However, these retention mechanisms are only effective when water passes through a large part of the soil pore space. When macropores (structural pores such as shrinkage cracks, worm and root channels) are present, infiltration mainly takes place as preferential flow, which occurs when vertical flow velocity is much higher than the lateral movement into the soil matrix (Jarvis, 1998). The residence time within the chemically and biologically more reactive topsoil may decrease drastically to minutes or hours instead of weeks or months due to preferential flow. In the subsoil, degradation and retardation processes are less intense, hence leaching of contaminants from structured soils to the groundwater considerably influence the effective half-lives of the components.

Colloid-facilitated transport causes a considerable increase in the mobility of low-solubility contaminants. For example, Ballard recorded in 1971 that the leaching of DDT (dichloro-diphenyl-trichloroethane) through forest-floor columns increased 30 times in the presence of humic acids compared to the control treatments. Understanding of the various processes that govern this mobility of solutes, e.g. colloid mobilization, adsorption of contaminants, transport and deposition is vital in the efforts to control leaching. The mechanisms of filtering and sedimentation, which normally play an important role, may be less effective in soils where macropore transport is important. This thesis was carried out to improve our knowledge of the extent to which colloid facilitated transport mechanisms operate in macroporous clay soils.

3.1 Aim

The objective of this study was to conduct lysimeter experiments to investigate the relationship between flow velocity and filtering of colloids in undisturbed soil columns, and to improve knowledge on how the macropore structure of a clay soil influences filtering and sedimentation processes.

3.2 Scope

First, a literature overview over the findings of colloid-facilitated transport is presented. Then the laboratory experiment is described and discussed. The study was restricted to clay colloid transport. Contaminants were not introduced in the experiment and will not be discussed thoroughly in the literature overview.

4. LITERATURE OVERVIEW

4.1 Preferential flow of water

Preferential flow may arise in clay soils as ***macropore flow***. Large structural pores that are vertically continuous transport the water rapidly downwards and lateral movement to the low-permeable dense matrix is too slow to spread the water (Jarvis, 1998). Preferential flow may also occur in other types of soils due to other mechanisms. Preferential ***finger flow*** may develop in texturally and structurally homogenous sands or in soils with a hydrophobic organic surface layer. Water accumulates on the surface and infiltrates initially at the points where the pressure head first overcomes the "water-entry" pressure, in surface depressions or where canopy-intercepted water reaches the surface. The water infiltrates as fingers instead of a uniform wetting front. A third case is ***funnel flow***, occurring in soils with a heterogeneous mixture of different grain sizes. At a given water pressure a certain pore size range is available, and among these the water will infiltrate the pores with the highest hydraulic conductivity. This report concentrates on macropore flow in clay soils.

A fast breakthrough in a lysimeter experiment implies that a small fraction of the macropores are active in the transport (Lægdsmand *et al.*, 1999). The larger the pores that the water can enter, governed by the water potential, the fewer the pores that will effectively participate in the transport. With a low flow intensity the water potential remains low and the largest macropores will not be filled. Preferential flow may occur without the largest macropores necessarily participating. As flow intensity increases, higher water potential allows the water to enter larger pores. Larger pores are less tortuous and have a smaller total pore wall area, making the contact and possible sorption to the walls less. The time for contact between suspended particles and the pore walls will of course also be less.

4.2 Solute transport processes

From the foregoing, it is clear that preferential flow may enhance the leaching of surface applied contaminants. The relative importance of preferential flow and matrix flow for solute transport differs according to the degree of sorption of the solute to soil. For very mobile soluble solutes (like nitrate), matrix flow will cause highest leaching amount related to input, since these agents are found within the soil matrix where the bulk of the resident soil water exists. For immobile, strongly sorbed solutes, preferential flow will cause most leaching (Brown *et al.*, 1995). Topsoil cultivation is a way to decrease preferential flow in structured clay soils, but this could at the same time increase the leaching of, for example, nitrate (Brown *et al.*, 1995).

Sorbing solutes are retained in the soil body only as long as the solid to which they are attached is not mobilized. Small fragments - colloids - of the solid phase that are detached in the solid-water interface may migrate considerable distances. The attached hydrophobic agents, which are low-soluble and therefore normally considered immobile, will through this colloid-facilitated transport become very mobile. The move-

ment of strongly adsorbed agents such as heavy metals (Pb, Cr, Cd, Ni, Cu and Zn), some pesticides (for example glyphosate, atrazine, DDT and PCB), radionuclides (U, Ra, Th, Pu and Am), PAH (polycyclic aromatic hydrocarbons) and the macro-nutrient phosphate, is largely governed by the transport characteristics of the colloids (Villholth *et al.,* 1997). For this transport, the surface properties, size and stability of the colloids are important.

4.3 Colloids - the carriers

Colloids are very small particles that, per definition, remain suspended in a liquid phase. Their size ranges between 1 nm and 10 μm (Stumm, 1992), separating them from smaller molecules and sedimenting particles. Colloids may be organic (detritus or bacteria) or mineralogical (clay or Fe-, Al-oxides) (Villholth *et al.,* 1997). Their proportionally large specific surface area and high density of functional groups makes them effective sorbents of low-solubility contaminants (Lægdsmand *et al.,* 1999).

Transportability of the colloids depends on if they can remain in suspension. Factors favouring suspendability are (Seta & Karathanasis, 1997):

- high repulsive potential (Φ) between colloids
  favoured by:
  - high ΔpH (difference between suspension-pH and pH_{PZC} of the colloids, clay colloids have their point of zero charge (PZC) at low pH, hence a high suspension-pH will increase the repulsive potential)
  - low ionic strength in the suspension (maintaining a thick diffuse double layer, DDL, around the colloids, which will keep them further apart than the reach of attracting van der Waals-forces)
  - adsorption of organic compounds on the colloid surfaces (increases surface charge and thus the repulsion)
  - high relative proportion of mono- to divalent cations in the bulk solution (keeping DDL thick)
  - low concentration of Fe and Al in solution (they enhance colloid aggregation by binding specifically to the colloid surface, neutralizing the surface charges)
- the shape of the particles (irregular surfaces provide less good contact between particles) (Oster *et al.*, 1980)
- low initial concentration of particles in the suspension (a high concentration increases the risk of flocculation)

4.4 Adsorption of the contaminants

Seta & Karatahanasis (1997) reported on factors enhancing adsorption of atrazine, a moderately strongly sorbed herbicide, on colloids. Increased levels in the colloid suspension of organic carbon, pH, CEC and total exchangeable bases favoured adsorption, whereas increased levels of kaolinite and Fe- and Al-oxides were inhibiting. Sorption of another, very strongly sorbed, herbicide, glyphosate, and its degradation

product AMPA, resembles that of inorganic phosphate (Villholth *et al.,* 1997). Fixation to the mineral fraction dominates, Fe- and Al-oxides at low pH and fixation by Ca at high pH, and only a small portion of the compound is mobile. The mobile fraction is largest between pH 6 and 7. Since the addition of inorganic P decreases glyphosate adsorption, a possible saturation of the soils with fertilizer P could render both inorganic P and glyphosate more mobile in dissolved form (Villholth *et al.,* 1997).

4.5 Mobilization

Colloids are mobilized either at the soil surface or within the soil body along the pore walls. Jarvis *et al.* (1999), assumed that surface mobilization is the most important source in shallow unsaturated soil profiles, and this is supported by other experiments. Hardy *et al.* (1999) concluded that 81 % of the particles in tile drainflow came from the uppermost 5 cm of the profile. On the soil surface, aggregates are broken down, and particles are detached and mobilized by the action of raindrops. The following equation expresses the rate of soil detachment, v_d, [g m^{-2} h^{-1}], on the soil surface:

$$v_d = k_d \, E_k \, [1 - \exp(-2Hg/64)] \, P_r \, M_s \qquad (1)$$

where k_d is the soil detachability coefficient [g J^{-1}], E_k is the kinetic energy [J m^{-2} mm^{-1}], H is the fall height [m], g is the acceleration of gravity [9.81 m s^{-2}], P_r is the rainfall intensity [mm h^{-1}] and M_s is the amount of readily available dispersible particles [g g^{-1} soil]. M_s depends on the rate of replenishment of readily available dispersible particles, of which little is known other than that it is influenced by ploughing, freezing and thawing, wetting and drying and mixing by soil biota (Jarvis *et al.*, 1999). The impact energy (E_k) is influenced by the fall velocity as well as the size of the droplets (*Jarvis et al.*, 1999), but can be expressed empirically as a function of rainfall intensity:

$$E_k = 29 \, [1 - 0.72 \exp(- 0.05 \, P_r)] \qquad (2)$$

The soil detachability coefficient (k_d) varies with organic matter content, the dominating clay minerals and cations present. Higher content of clay, organic matter and water are factors increasing the total amount of potentially dispersible particles, as well as physical surface properties like crusts (Jarvis *et al.*, 1999). It is at the beginning of a rain event that particles are most frequently mobilized (Grant *et al.*, 1996). Particles may be detached due to dispersion when the ionic strength around the aggregates is lowered by the rainwater dilution (Jarvis *et al.*, 1999). Rapid wetting of dry soil causes physical stress, which may also lead to aggregate breakdown.

In macroporous soils, physical perturbation is the most common form of internal colloid mobilization (Ryan & Elimelech, 1996). Hydrodynamic shear stress caused by acceleration of the flowing water moving into smaller channels leads to mobilization. One would expect that a greater mobilization would occur at a higher flow rate due to a higher hydraulic stress on the macropore wall. Yet, Ryan and Gschwend (1994 a) found that the release of particles decreased with increasing flow rates and suggested that the reason was rate limited diffusion of detached colloids across a boundary layer

between grain surfaces and the bulk fluid. Increasing flow rates also mean that larger macropores are in use, and the pore wall area in contact with the water will be less.

Different kinetics control the rate at which the different particle sizes are released from the macropore wall (Jacobsen *et al.*, 1997). Small particles are adsorbed more strongly to the macropore wall due to their relatively large surface charge. Larger particles are more exposed to hydraulic forces.

Reducing the ionic strength of the suspension promotes colloid mobilization, since the diffuse double layer around the colloids grows and makes them repellent. Lowering the suspension pH may dissolve organic cementing agents, and a change of redox potential may dissolve the cementing Fe-oxide goethite (Ryan & Gschwend, 1994 b). In contaminant plumes, which are often characterized by high ionic strength and low pH, colloid mobilization may not seem likely, since these properties are the reverse of what is favourable. However, contaminant plumes often contain other compounds, such as organic acids, organic macromolecules, reductants and detergents, capable of causing enhanced colloid mobilization through reversal of surface charges or dissolution of cementing mineral phases (Ryan & Elimelech, 1996).

4.6 Filtering

Filtering includes physical sieving by pore necks, physiochemical interactions with the charged soil matrix, gravitational settling and sedimentation due to decreased velocity in pore curves (McDowell-Boyer *et al.*, 1986).

Large particles are often sieved in pore necks and are more subjected to sedimentation. Small particles are delayed in the tortuous paths of the smallest pores. Consequently the medium sized particles (the size depending on the pore size distribution of the soil) have the highest mobility (Villholth *et al.*, 1997). The mass recovery is higher with increasing flow rate, and a higher proportion of large particles is leached, because at higher intensities larger macropores participate in the transport, where there is less sie-ving and sedimentation (Jacobsen *et al.*, 1997).

Seta and Karathanasis (1997) found that a higher applied colloid concentration yields higher recovery through the lysimeters. They had several explanations: a higher concentration gives decreased colloid stability, i.e. promotes coagulation. This formation of larger particles make them physically excluded from small pores, which result in a straighter path. There will be greater saturation of colloid binding sites on the macropore wall. Colloid-binding to the walls is only monolayered.

Filtering may be calculated as follows:

$$F = f_c \, c \, q \qquad (3)$$

where F is the rate of filtering of particles [g m^{-3} h^{-1}], f_c is the filter coefficient [m^{-1}], c is the concentration of suspended particles [g m^{-3}] and q is the water flux [m h^{-1}]. The

filter coefficient, f_c [m^{-1}], can be calculated from the results of a soil column breakthrough experiment:

$$f_c = - \ln (c_{out} / c_{in})_{plateau} / z \quad (4)$$

where c_{out} is the concentration of the outflow suspension at the plateau of the breakthrough curve, c_{in} is the concentration of the input suspension and z is the depth [m] of the soil column.

4.7 Models

Many simulation models have been developed to predict the fate and pathways of contaminants through the soil, but most deal only with flow in homogenous soil columns. In recent years, a few models dealing with preferential flow in macropores have been developed, for example the RZWQM model (root-zone water quality model) (Ahuja *et al.*, 1999) and the MACRO model (Jarvis, 1998) . However, when it comes to colloid-facilitated transport there are a few well-developed models for homogenous soils, but much remains to be understood of the colloid transport processes occurring in structured heterogeneous soils.

5. MATERIALS AND METHODS

Four replicate soil monoliths with a diameter of 30 cm and a depth of 0.5 m were sampled in August 1997 in a closely spaced row at the site "Ultuna 1", which is a field used for cereal cropping situated 6 km south of Uppsala (60°N, 17°E), Sweden. The soil is a post-glacial clay loam, overlying a varved glacial clay loam, classified as a Typic Eutrochrept (Eriksson, *pers. com.*, 1999) according to the USDA Soil Taxonomy. More detailed soil data is given in Appendix 1.

The soil monoliths were carefully drilled out and pushed into plastic cylindrical casings without altering the soil structure. The drilling technique is described by Persson & Bergström (1991). Lids were placed at the bottom and the lysimeters were stored in darkness at 4 °C. After carefully removing 1 cm of soil at the bottom, without any visually detectable smearing of the surface structure, a coarse filter (0.3 mm pores) and a plastic plate were placed inside the bottom lid. The plate had depressions channelling the water to a pair of outflow tubes passing through the lid. The filter prevents coarse material from entering the tubes. The dead-volume is approximately 450 ml, including tubes. The lysimeters were slowly (in order to avoid air getting trapped) saturated from the bottom with artificial rain water with pH 5,5 and conductivity 26 μScm^{-1} (see Table 1 for composition of the rain water) and then gravitationally drained to reach equilibrium with a water table located at the base of the column (at 50 cm depth).

Table 1. Composition of artificial rain water (Lægdsmand *et al.*, 1999)

Salts	Concentration (mmol/l)	Concentration (mg/l)
$(NH_4)_2SO_4$	0.030	3.41
$CaCl_2 \times 2\ H_2O$	0.0112	1.65
KCl	0.0047	0.35
$MgSO_4 \times 7\ H_2O$	0.0121	2.98
NaCl	0.056	3.24
$NaNO_3$	0.048	4.07

Colloids smaller than 2 μm in diameter were isolated from the Ultuna 1-soil through sedimentation fractioning. The extraction was accomplished by mixing 20 g soil in 500 ml deionized water in a propeller-mixer for 40 minutes, repeating the procedure in order to get enough material for the experiments, and letting it sediment. According to Stoke's Law, particles larger than 2 μm (silt fraction) sediment 10 cm in 8 hours, hence after 24 hours the upper 30 cm suspension was collected with a siphon. Stokes' law describes the relationship between sedimentation velocity and particle size:

$$v = 2\ r^2 (\rho_s - \rho_l)\ g\ /\ (9\ \eta) \qquad (5)$$

where v is the constant sedimentation velocity [cm s^{-1})], r is the equivalence radius of the particle [cm)], ρ_s and ρ_l are the particle and liquid densities [g cm^{-3}], respectively, g is the acceleration due to gravity [m s^{-2}] and η is the viscosity of the liquid [g cm^{-1} s^{-1}]. The clay colloids were then suspended in artificial rainwater to a concentration of 1 g/l. The correlation between turbidity (light extinction) and mass weight was then determined. A dilution sequence with 3 $\times$ 2 replicates was obtained: 3 replicates of the diluting (pipetting) step, 2 replicates of the pipetting of an exact volume. By drying the volume samples at 105 °C overnight to obtain the dry weight, the exact concentration could be determined. Turbidity was measured on the stock solutions of each dilution step (i.e. 3 replicates) with a Hach Model 2100N Turbidimeter. Every sample was shaken thoroughly, air bubbles were given 20 seconds to disappear, and the turbidity was measured several times with a gentle shaking of the cell in between. A calibration curve was constructed by plotting the means of the turbidity measurements against the corresponding mean concentrations (see Figures 1 and 2):

$$c = (t + 743.04) / 2374{,}8 \qquad (6)$$

where c is the colloid concentration [g L^{-1}] and t is the turbidity [NTU]. This calibration function was not used in the analysis of the results, for reasons which are discussed below.

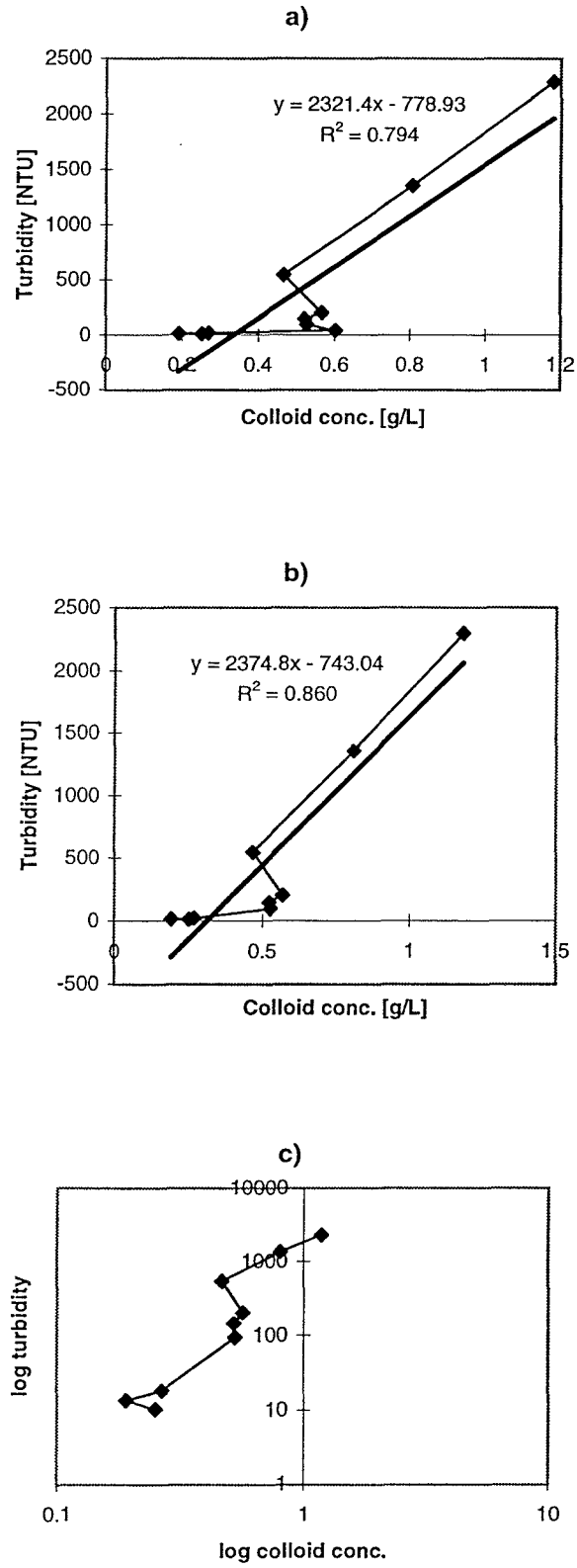


Figure 1. Standard function diagrams. a = Standard function with all average data points (see Appendix 2). b = Standard function without one extreme data point (see Appendix 2). c = log-log-diagram of the standard function without one extreme data point.

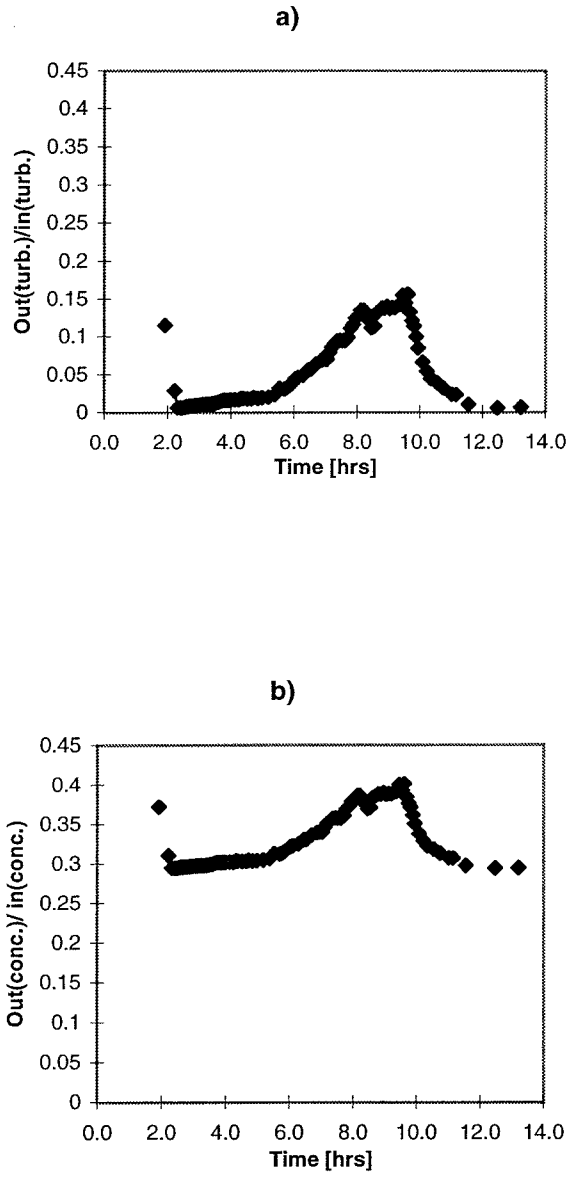


Figure 2. Breakthrough curves for lysimeter B (irrigation intensity 5 mm/h) illustrating the conversion from turbidity to concentration. a = = Breakthrough curve plotted from turbidity data. b = Breakthrough curve plotted from concentration calculations.

The irrigation equipment used to apply the colloid solutions to the soil columns is shown in Figure 3 together with the sample collection equipment. It comprises:

1) A bucket containing 2 l of either artificial rainwater (for initial "flushing" of detachable soil colloids) or colloid-rain suspension. The speed of the propeller mixer keeping the suspension homogenous was 750 rpm. A mechanical lifter was used to compensate for the falling water surface in the bucket.
2) The water was evenly spread over the lysimeter surface with 36 capillary tubes. A level tube showed the current water pressure driving the application. The droplets emerged 4-6 cm above the soil surface.
3) Open contact with atmospheric pressure before the outflow water enters the sampling tube. At flow intensities exceeding the sampling intensity of the pump (4), water continued to the overflow drainage.
4) A sample pump. At a rain intensity of 5 mm/h, the speed of the pump exceeded the outflow velocity, whereas the intensities 9 and 13 mm/h resulted in overflow drainflow.
5) Automatic sample collector. Maximum sample volume was 34 ml, equivalent to 0.50 mm of drainage.

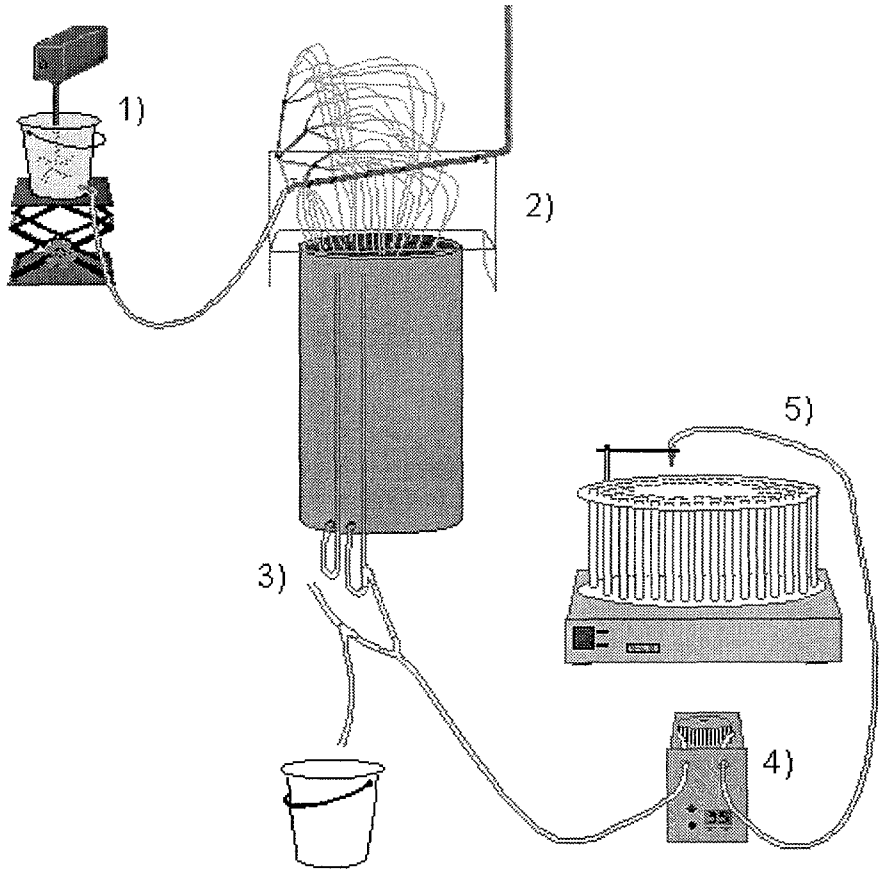


Figure 3. Irrigation equipment and sample collection equipment. 1 = bucket containing input liquid. 2 = irrigation device. 3 = open contact with atmospheric pressure. 4 = sample pump. 5 = sample collector. For further explanations, see text.

The following irrigation intensities were applied to the four lysimeter replicates A, B, C and D: 9 mm/h on lysimeters A and D, 5 mm/h on B and 13 mm/h on C. The intention had been to run several intensities on each lysimeter, but the extended rain seemed to induce loss of aggregate stability and internal erosion, so that only the first data series was reliable. Each run consisted of 3 phases:

phase 1: measuring internal erosion by applying only artificial rain water.
phase 2: external input of colloids by applying a colloid-rain-suspension
phase 3: recession, where the rain application was instantly turned off but samples were collected as long as there was an outflow.

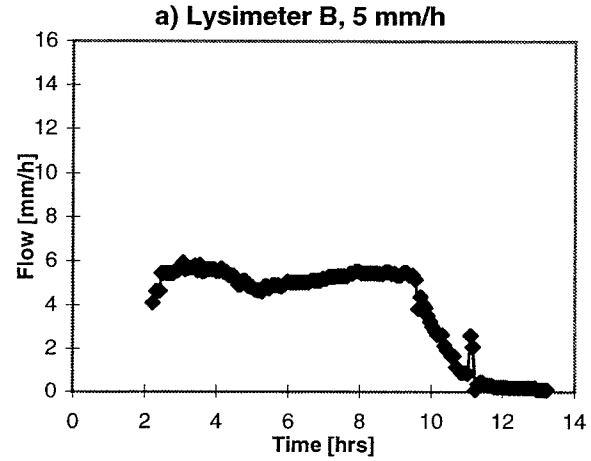
There was a 4 - 8 minutes break of the input between the end of phase 1 and start of phase 2 when the rain water was replaced by the colloid-rain-suspension .

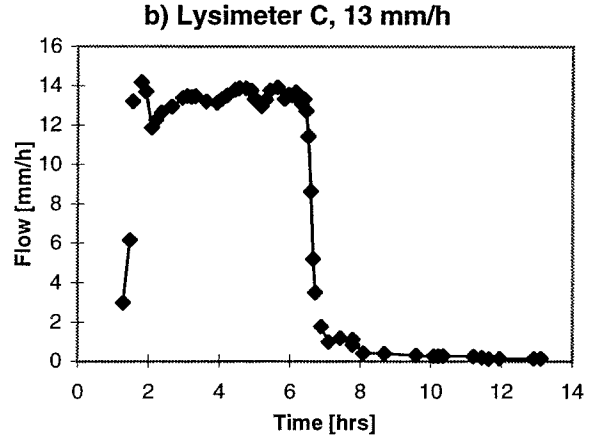
Outflow liquid samples were collected continuously with a pump, with 3-5 minutes outflow collected per sample depending on flow velocity. Maximum sample volume was 28 ml, equivalent to 0.41 mm of drainage. Flow velocities of the effluent water were also registered for total flow calculations. For lysimeter B, irrigation intensity of 5 mm/h, the flow velocity was measured as the volume of each sample. For lysimeters C, 13 mm/h, and D, 9 mm/h, the flow rate exceeded the sampling pump rate, and the overflow drainflow rate was registered by taking the change in water mass between two points of time, approximately every 5 minutes. The colloid concentrations in the collected samples were analyzed on the turbidimeter. The minimum sample volume for the cuvette was 17 ml. The samples were shaken immediately before measuring. The effluent concentration was compared with an average value over the whole application period of the influent concentration. Conductivity and pH was also measured in the artificial rain and in the influent colloid suspensions.

6. RESULTS AND DISCUSSION

6.1 Water flow

Figures 4a to 4c shows the water flow recorded for lysimeters B, C and D. For lysimeter B the average maximum flow was 5.3 mm/h, for lysimeter C it was 13.2 mm/h. The flow diagram of lysimeter D shows the very irregular outflow pattern that this lysimeter exhibited. This behaviour could imply that air had been trapped in the system, possibly due to insufficient water saturation of the lysimeter, or air bubbles in the tubes.





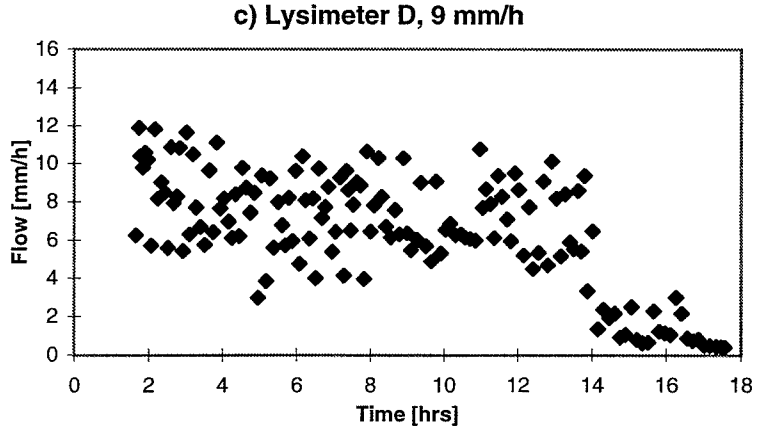


Figure 4. Water flow in effluent. a = lysimeter B, irrigation intensity 5 mm/h. b = lysimeter C, irrigation intensity 13 mm/h. c = lysimeter D, irrigation intensity 9 mm/h.

According to calculations using pressure head data in Table 3, Appendix 1, the total water-filled pore volume in the lysimeters at drainage equilibrium was 193 mm (≈13 dm^3). This includes some contribution from matric pores. However, in the experiment there was a rapid increase in the output flow rate after the lag in phase 1, and this together with the rapid recession in phase 3 is a clear indication of macropore flow. If the limit for macropores is set to pores larger than 0.6 mm, which corresponds to when hydraulic conductivity starts to increase rapidly with pore size, according to measurements made by Messing (1993) on the Ultuna soil, the volume of the conducting macropores will be 14 mm (≈ 0.96 dm^3), or 0.03 m^3 m^{-3} soil. This lower value represents the active conducting pore volume, and also the amount of water required to move a mobile solute through the column. This value must be considered highly theoretical, since natural variations in the structure are very likely. One single large crack would have a great influence on the hydrology of the column due to the high conductivity of the crack. One indication of difference in macroporosity between soil replicates was the water breakthrough time, from which the macroporosity may be calculated:

$$t = z\ \theta / q \qquad (7)$$

where t is the breakthrough time [h], z is the depth [mm] of the soil column, θ is the conducting porosity and q is the flow velocity [mm/h]. The estimated active macroporosity varied between less than 1 % (lysimeter B) to more than 3.5 % (lysimeter C). These estimates were in good agreement with the estimates made from static pressure head data above.

6.2 Particle transport

The particle transport analyses are presented as breakthrough curves with the effluent fraction of the influent colloid turbidity plotted as a function of time (see Figure 5). The derived standard curve (Figure 1b) was considered useless for converting the turbidity measurements to concentrations . Firstly, the function did not pass through the origin, resulting in a function constant moving the base level of the concentration curve upward more than the amplitude of the whole curve. Secondly, there was a clear tendency in the standard series that more colloids, measured by weight, were contained in the samples than was representative for a certain dilution step. Turbidity, on the other hand, was lower than expected for each dilution step. The proportions between particle sizes might not have been constant throughout the dilution series. Some size fraction might have been over-represented in the pipetting-steps, due to insufficient mixing of the suspension and the fact (despite the use of a propeller mixer) that particles may adsorb to surfaces. It is possible that small particles influence turbidity more than the weight of the sample, so that turbidity does not accurately represent colloid concentration. The relationship between weight and turbidity might not be linear for colloids. Moreover, humic substances might have another relationship between colour and weight than mineral colloids, causing an overestimation of turbidity in samples with low colloid concentration. Spectrophotometry was used by for example Jacobsen *et al.* (1997), while Lægdsmand *et al.* (1999) used turbidity. Different particle sizes and minerals may in spectrophotometry absorb varying amounts of different

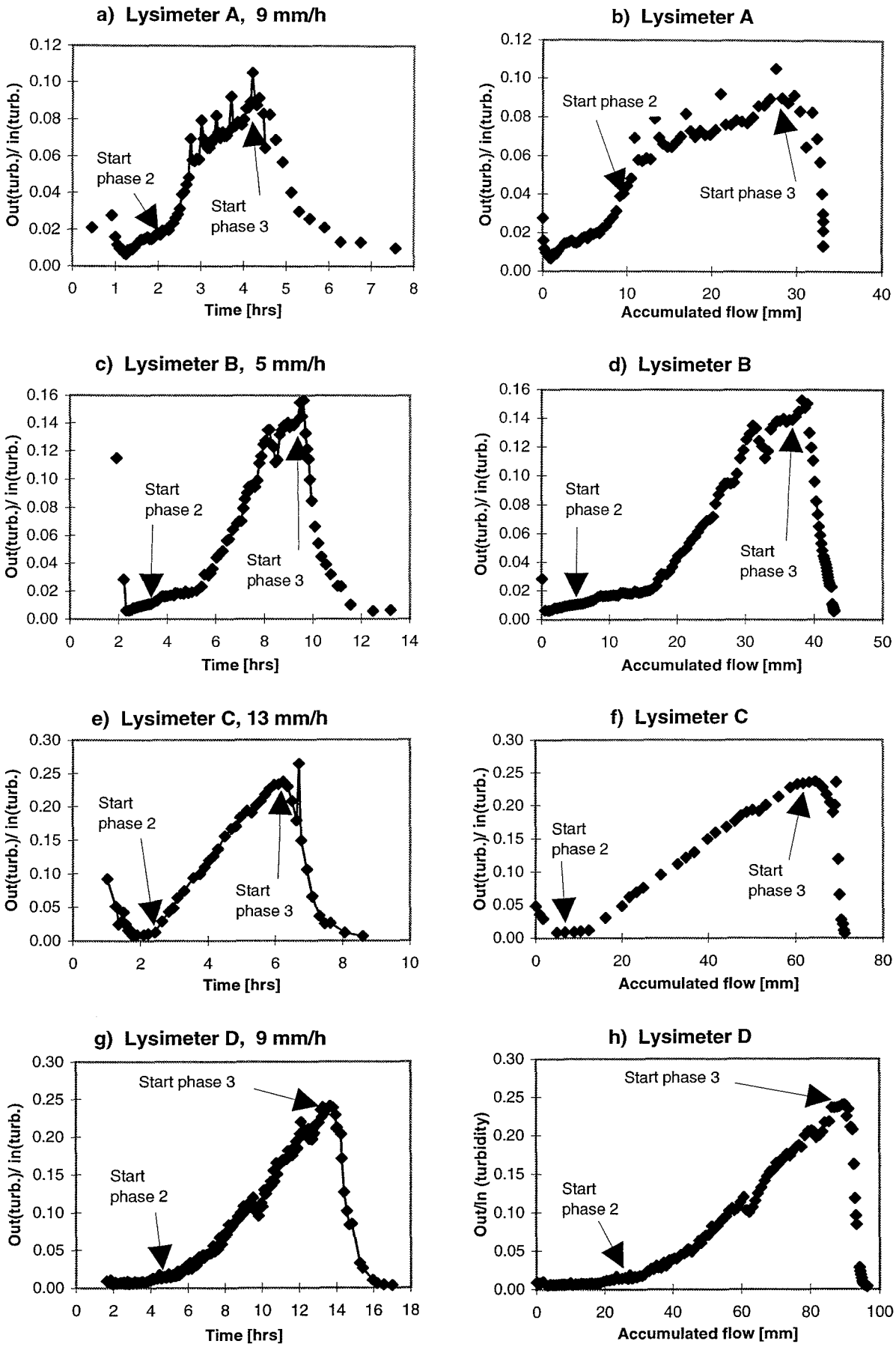


Figure 5. Breakthrough curves for lysimeter A (a, b), lysimeter B (c, d), lysimeter C (e, f) and lysimeter D (g, h). Attention! Different scales.

wavelengths, and at low wavelengths (<700 nm) light absorption by organic matter becomes significant (Wilander, *pers. com.*, 1999).

The first data points of phase 1 represent an average of the effluent concentrations before the flow velocity exceeded the minimum analysis volume of the turbidity cuvette, i.e. 17 ml/3 min. for lysimeters A and D, 17 ml/6 min. for lysimeter B and 17 ml/5 min. for lysimeter C. Possible high initial concentrations of internally generated colloids in the first few effluent drops may therefore have been diluted by later effluent of lower concentration. After the first pulse of colloids, the turbidity decreased to very low levels before it began to rise again (Figure 5). We had expected a continuous base flow of internally generated colloids throughout the experiment, that would have had to be subtracted from the breakthrough curve, but instead there was an initial flush of the loose particles in the macropores, and a base flow was then undetectable, probably due to low internal mobilization rate.

The slope of the breakthrough curve in phase 2 was almost twice as steep for intensity 13 mm/h as for intensity 5 mm/h (Figure 6). A linear function adjusted to the sloping part of phase 2 show a slope of 0.034 for intensity 5 mm/h and a slope of 0.061 for intensity 13 mm/h. The slope is the increase in relative colloid output. A possible interpretation is that with a higher rain intensity fewer, but larger, pores are engaged which gives less total pore wall area, fewer binding sites and lower filtration.

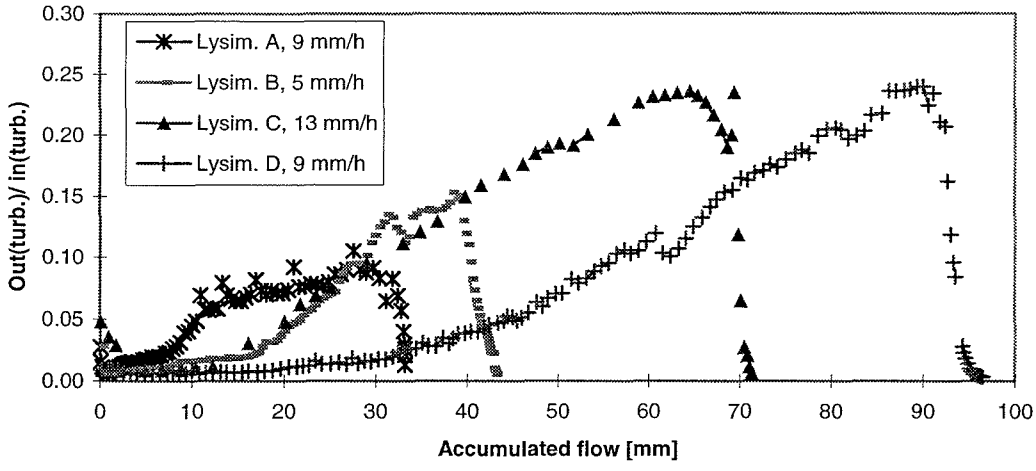


Figure 6. Breakthrough curves from lysimeters A, B, C and D.

Theoretically it takes one pore volume to move a mobile solute through a soil column. At one pore volume, the output concentration of a mobile solute has in theory reached half its maximum value. We had expected a rapid increase of the relative output of colloids up to a maximum level or plateau, determined by filtering efficiency. However, there was no sign of a plateau at two pore volumes after the start of colloid application in any of the lysimeters, and not later either, despite the fact that phase 2 was run more than four times as long as the expected breakthrough time for a mobile solute for lysimeter C and D. There may have been some additional colloid source

masking the breakthrough of the externally added colloids. It could be internal erosion or originate from surface impact mobilization. One hypothesis was that the water used for saturation of the monoliths had too weak ionic strength - we used the artificial rain water, which had an ionic strength of 0.3 mM - and that soil drainage water with an ionic strength of approximately 7 mM (Wiklander & Andersson, 1974) would have been better. The lower part of the monoliths, accustomed to soil water with high ionic strength, may have been subject to a dispersive weakening process, which made them susceptible to internal erosion. Mobilization was accelerated as more water passed through. However, this idea is contradicted by the breakthrough curves which show that the colloid output ceased between the first "internal erosion-peak" in phase 1 and the increase in phase 2 (Figure 5).

Another explanation could be that raindrop impact on the surface - although the fall height was merely 5 cm on average - may have caused mobilization of colloids. The rain application was distributed on 36 points, and although these were moved every 20 minutes, erosion due to the concentrated flow on these spots may have occurred. This hypothesis that surface erosion lay behind the increase in turbidity is contradicted by two circumstances: firstly, the fact that the output turbidity remained high even when textile wicks were used to apply the droplets avoiding drip impact. Secondly, the idea that there is a limited source of readily available dispersible colloids, and that the rate of replenishment of this source is too slow for the time scale of this experiment. This is supported by the fact that the colloid output did cease in phase 1. Maybe surface erosion occurred for a short period, and lay behind the turbidity peak in phase 1, but was not the reason behind the continuing increase in turbidity in phase 2.

A third hypothesis was that the colloid-rain-suspension had an unfavourable chemical composition, but this is contradicted by the fact that the colloid-suspension in phase 2, having a slightly higher pH and ionic strength than the artificial rain in phase 1, would, according to the literature (Ryan & Gschwend, 1994 b; Ryan & Elimelech, 1996), have made the aggregates stronger. This still leaves the rising turbidity in phase 2 unexplained.

A fourth idea is that turbidity is affected by the particle size distribution in the colloid suspension. As discussed earlier, small colloids seem likely to exhibit more turbidity than weight, compared to larger colloids. If we assume that the breakthrough curves of this study (Figure 5), with a slowly rising turbidity with time, would show a more rapid increase if expressed as concentration against time, and would exhibit the "normal" breakthrough shape, a plausible explanation would be that the pore size distribution of the effluent is changed with time towards a greater fraction of smaller particles. There is a possibility that the particle size distribution is changed through the soil column, due to selective filtering of certain particle sizes. The distribution might also be changed with time in the effluent, as a result of outflow concentrations representing flow pathways of different transit times. At first larger particles dominate in the outflow, causing a rapid increase in concentration but a more moderate increase in turbidity. Later the proportion of smaller colloids in the particle size distribution is increased, and the concentration curve levels out while the turbidity curve still rises.

In this experiment, the variation between soil replicates could not be eliminated, since only one application intensity was run on each lysimeter. The intention was, initially,

to compare different application intensities on each lysimeter, but the lysimeters could not be flushed of remaining colloids between runs. The outflow turbidity did not decrease even after extended flushing. This tends to support the first explanation given above for the increase of turbidity with time in the outflow.

There is a possibility that the plateau would have been reached if phase 2 in the experiment had continued longer. This would mean that dispersion was considerable, since the rising limb of the breakthrough curve would have this slow increase. It would imply that colloids were delayed in smaller pores, and thus preferential flow of particles did not dominate the particle transport, but particles did enter the soil matrix. This is contradicted by the steep rising and recession limbs of the water breakthrough curve (Figure 4), which is clear evidence of preferential macropore flow.

6.3 Filtering

The maximum outflow turbidity reached 25 % of the applied turbidity before the rain application was stopped (Figure 6). Due to the additional colloid source, the value might have increased further if the experiment had continued. It is difficult to say anything about the filtering percentage since we do not know the size of any additional colloid source. Nevertheless, if we assume a plateau after two pore volumes, and a theoretical pore volume of 14 mm, the leaching would in theory be 10 % in lysimeter B (5 mm/h) and 14 % in lysimeter C (13 mm/h). This would imply that filtering is proportionally less effective at a higher flow intensity.

Using Equations 1 and 2, the amount of colloids mobilized by surface erosion during the experiment was calculated. According to calculations, 54 g m^{-2} h^{-1} colloids could be mobilized at a rain intensity of 13 mm/h. At a rain intensity of 5 mm/h, 15 g m^{-2} h^{-1} could theoretically be mobilized. This is the maximum rate of mobilization, but as the pool of readily available dispersible particles decreased, the limiting factor was the rate of replenishment. Approximately 3 g were totally applied with the colloid-rain-suspension.

Since no clearly defined plateau was reached in any of the breakthrough experiments, no real filter coefficient could be calculated, and the variation of filtering with flow velocity could not be properly investigated, which was the aim of this study. Nevertheless, assuming the imaginary plateau level after 2 pore volumes, lysimeter B (rain intensity 5 mm/h, Figures 5c & 5d) would theoretically have a filter coefficient of 4.6 m^{-1} and lysimeter C (13 mm/h, Figures 5e & 5f) a value of 3.9 m^{-1}. The values are of the same order of magnitude as others have reported. Jarvis *et al.*(1999) found a best-fit value of the filter coefficient of 2 m^{-1} for a silty clay soil, and Jacobsen *et al.* (1997) reported filter coefficients ranging from 0.5 to 8 m^{-1}. The interpretation of these values, although they lack both verification and correspondence in the experiments, is that higher rain intensities would cause a lower filtering, and this agrees well with the idea that higher flow intensities engage larger pores, which are straighter and have fewer pore necks.

7. CONCLUSIONS

To conclude, leaching of 25 % of applied colloids is a high percentage. The theoretical values of 10 % and 14 % at the imaginary plateau are still high values, in the perspective of contaminants leaching to groundwater. The rapid increase of the additional colloid source was also alarming, if it originated from the surface, considering that pesticides are surface-applied and air pollutants are deposited on the surface, all easily being subjected to colloid-facilitating transport. The most plausible explanation for the additional colloid source is a combination of the idea that the soil monoliths did not endure the saturation with water of low ionic strength, and the dispersive process was accentuated during the experiment, and secondly the idea that turbidity did not uniquely represent the colloid concentration of the effluent, but varied with particle size distribution.

There is a need to develop the methodology for working with colloid suspensions, since they are non-homogenous and difficult to obtain representative samples from, in terms of particle size distribution.

Another suggestion for further research is that the added colloids are marked so that they may be separated from internal colloids. This, of course, requires money and equipment for detection. It would be desirable to design an experiment where surface-mobilized colloids (the uppermost 3-5 cm) are marked, so that surface erosion and internal erosion may be measured as two separate fractions. This could be accomplished by analysing for radioactive Cs from natural fall-out which is very strongly sorbed to particles on and near the surface.

8. ACKNOWLEDGEMENTS

I would like to thank my supervisor professor Nicholas Jarvis for enlightening discussions on soil physics and for your inexhaustible optimism throughout the project. Lave Persson, my indispensable technical support, thank you for your interest and enthusiasm ('engagemang' in Swedish) and your many fruitful ideas. Petra Bergkvist, my second supervisor, thank you for your great support in the practical laboratory work, and for formulating many fruitful questions for me to answer. Last but not least, I would like to thank Joakim Elgh for supporting me in many ways throughout the process.

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APPENDIX 1. Physical and mineralogical properties of the Ultuna 1 soil.

Table 1. Particle size distribution (weight %) of the Ultuna 1 soil (Wiklert *et al.*, 1969)

Depth (cm)	Clay	Silt	Sand	Organic content
0-10	37	43	20	1
10-20	38	42	20	1
20-30	38	43	19	1
30-40	38	42	20	1
40-50	37	43	20	0

Table 2. Physical properties of the Ultuna 1 soil (Wiklert *et al.*, 1969)

Depth (cm)	Physical wilting point (vol. %)	Compact density ($g\ cm^{-3}$)	Dry bulk density ($g\ cm^{-3}$)	Hydraulic conductivity, k ($cm\ h^{-1}$)	pH (H_2O)
0-10	16	2.65	1.36	8.5	6.5
10-20	20	2.71	1.50	1.7	6.7
20-30	19	2.69	1.49	14	7.0
30-40	23	2.69	1.57	34	7.2
40-50	19	2.66	1.44	83	7.5

Table 3. Relationship between water content and pressure head for the Ultuna 1 soil (Wiklert *et al.*, 1969)

Depth (cm)	Porosity (%)	Water content (% v/v) at a pressure head (m w c) of 0.05	0.15	0.50	1.00	4.00	10.00	50	150 at sampling
0-10	49	46	42	37	36	32	31	26	15
10-20	45	40	38	37	36	34	22	28	14
20-30	45	43	39	38	37	34	33	30	19
30-40	42	42	38	36	34	32	31	29	24
40-50	46	42	35	33	33	31	29	27	19
									20
									25
									18
									24

Table 4. Mineralogical composition of three particle size fractions (μm) of the Ultuna soil (Ghorayshi & Öborn, 1993)

Fraction	Mi[1]	Sm[1]	V[1]	Ch[1]	K[1]	Q[1]	Kf[1]	Pl[1]	Am[1]
<0.2	++++++[2]	0	+	0	+	0	0	0	0
0.2-2	++++	0	+	t	+	++	++	++	+
2-20	+	0	+[3]	t	t	++	+++	+++	+

[1]) Mi = mica (illite); Sm = smectite; V = vermiculite; Ch = chlorite; K = kaolinite; Q = quartz; Kf = K-feldspars; Pl = plagioclases; Am = amphiboles.

[2]) 0 = not traceable; t = trace (≤ 2 %); + = >2 % and < 10 %; ++ = 10-20 %; +++ = 20-30 %; ++++ = 30-40 %; +++++ = 40-50 %; ++++++ = more than 50 %

[3]) Includes vermiculite with hydroxyaluminium interlayers.

APPENDIX 2. Standard function data.

Table 1. Concentration and turbidity data constituating the standard function (see Eq. 6)

Each row in columns 1 and 4 represents averages of 2 repetitions of a dilution step. Columns 2 and 3 are averages of every three rows in column 1 and 4, respectively.

	To diagram:	To diagram:	
Conc.	**Aver. conc.**	**Turbidity**	**Turbidity**
[g/L]	[g/L]	[NTU]	[NTU]
0.2262	0.2518	10.2	10.5
0.2746	0.1917	13.5	9.95
0.2547	0.2683	18.1	10.2
	~~0.6027~~	~~41.6~~	
0.1908	0.5276	94.1	13.5
0.1836	0.5225	145.2	13.6
0.2008	0.5678	203.0	13.5
	0.4675	546.0	
0.2292	0.8072	1355.3	18.0
0.2912	1.1820	2294.0	18.3
0.2844			~~14.0~~
0.5449			41.6
0.6403			41.0
0.6228			42.2
0.5375			93.8
0.5230			94.4
0.5224			94.2
0.5296			144
0.5252			145
0.5128			147
0.5475			199
0.5443			203
0.6115			206
0.4905			540
0.4530			550
0.4590			548
0.7720			1361
0.8005			1355
0.8490			1350
1.1820			2294
1.9445			Too high

APPENDIX 3. Water flow data and breakthrough turbidity data

Lysimeter A								
Time	Tot.outflow	Time	Turbidity	Time	Accum. flow	Out/In (turb.)		
(h in dec.fc	[mm/h]	(h in dec.fc	[NTU]	(h in dec.fc	[mm]			
0.93	1.76	0.47	39.57	0.87	0.03			
1.01	5.27	0.93	51.83	0.87		0.0276		
1.06	7.46	1.01	29.90	0.98	0.11	0.0000		
1.11	8.82	1.06	21.98	0.98		0.0159		
1.16	9.44	1.11	18.10	1.03	0.24	0.0000		
1.21	9.70	1.16	17.84	1.03		0.0117		
1.26	9.88	1.21	13.20	1.08	0.40	0.0000		
1.31	9.88	1.26	12.30	1.08		0.0097		
1.36	9.92	1.31	16.60	1.13	0.58	0.0000		
1.41	9.88	1.36	17.25	1.13		0.0095		
1.46	9.88	1.41	17.05	1.18	0.77	0.0000		
1.51	9.98	1.46	19.48	1.18		0.0070		
1.56	9.92	1.51	22.52	1.23	0.97	0.0000		
1.61	9.95	1.56	24.43	1.23		0.0066		
1.66	9.79	1.61	26.98	1.28	1.18	0.0000		
1.71	9.81	1.66	27.25	1.28		0.0089		
1.76	9.82	1.71	28.32	1.33	1.40	0.0000		
1.81	9.96	1.76	29.64	1.33		0.0092		
1.86	10.01	1.81	27.75	1.38	1.63	0.0000		
1.91	9.92	1.86	27.42	1.38		0.0091		
1.96	9.88	1.91	28.85	1.43	1.87	0.0000		
2.01	9.92	1.96	32.18	1.43		0.0104		
2.06	9.88	2.01	33.40	1.48	2.11	0.0000		
2.11	9.83	2.06	31.94	1.48		0.0120		
2.16	9.83	2.11	36.48	1.53	2.37	0.0000		
2.21	9.81	2.16	36.14	1.53		0.0130		
2.26	9.88	2.21	37.58	1.58	2.63	0.0000		
2.31	9.92	2.26	36.82	1.58		0.0144		
2.36	9.88	2.31	42.28	1.63	2.90			
2.41	9.88	2.36	44.00	1.63		0.0145		
2.46	9.92	2.41	49.10	1.68	3.17			
2.51	9.96	2.46	52.78	1.68		0.0151		
2.56	9.98	2.51	58.47	1.73	3.46			
2.61	9.92	2.56	73.23	1.73		0.0158		
2.66	9.99	2.61	75.73	1.78	3.75			
2.71	9.96	2.66	83.18	1.78		0.0148		
2.76	10.01	2.71	90.52	1.83	4.06			
2.81	9.92	2.76	130.00	1.83		0.0146		
2.86	9.88	2.81	108.25	1.88	4.37			
2.91	9.92	2.86	107.25	1.88		0.0154		
2.96	9.88	2.91	110.00	1.93	4.69			
3.01	9.83	2.96	109.00	1.93		0.0172		
3.06	9.83	3.01	148.67	1.98	5.02			
3.11	9.81	3.06	130.00	1.98		0.0178		
3.16	9.88	3.11	124.00	2.03	5.35			
3.21	9.92	3.16	121.00	2.03		0.0170		
3.26	9.88	3.21	121.00	2.08	5.69			
3.31	9.94	3.26	126.00	2.08		0.0195		
3.36	9.92	3.31	131.25	2.13	6.04			
3.41	9.96	3.36	153.25	2.13	0.00	0.0193		
3.46	9.98	3.41	136.75	2.18	6.40			
3.51	9.92	3.46	131.00	2.18	0.00	0.0200		

APPENDIX 3. Water flow data and breakthrough turbidity data

Lysimeter A continued								
Time	Total outflov	Time	Turbidity	Time	Accum. flow	Out/In (turb.)		
(h in dec.fc	[mm/h]	h in dec.forr	[NTU]	(h in dec.fc	[mm]			
3.56	9.88	3.51	137.00	2.23	6.77			
3.61	9.92	3.56	132.75	2.23	0.00	0.0196		
3.66	9.88	3.61	133.00	2.28	7.14			
3.71	9.83	3.66	138.00	2.28	0.00	0.0226		
3.76	9.83	3.71	172.50	2.33	7.53			
3.81	9.81	3.76	142.75	2.33	0.00	0.0235		
3.86	9.88	3.81	142.75	2.38	7.92			
3.91	9.92	3.86	147.00	2.38	0.00	0.0262		
3.96	9.88	3.91	145.75	2.43	8.32			
4.01	9.94	3.96	144.25	2.43	0.00	0.0282		
4.06	9.92	4.01	149.75	2.48	8.73			
4.11	9.90	4.06	160.75	2.48	0.00	0.0312		
4.16	9.94	4.11	161.25	2.53	9.16			
4.21	9.88	4.16	167.50	2.53	0.00	0.0391		
4.26	9.92	4.21	197.00	2.58	9.58			
4.31	9.92	4.26	168.25	2.58	0.00	0.0404		
4.36	9.88	4.31	163.75	2.63	10.02			
4.40	9.83	4.36	171.00	2.63	0.00	0.0444		
4.51	9.66	4.46	155.25	2.68	10.47			
4.61	9.04	4.51	120.75	2.68	0.00	0.0483		
4.71	7.46	4.61	154.50	2.73	10.92			
4.92	4.65	4.76	129.00	2.73	0.00	0.0693		
5.11	2.55	4.92	106.20	2.78	11.38			
5.31	1.23	5.12	75.28	2.78	0.00	0.0577		
5.56	0.57	5.31	55.75	2.83	11.85			
		5.56	48.60	2.83	0.00	0.0572		
		5.91	39.40	2.88	12.33			
		6.28	24.74	2.88	0.00	0.0587		
		6.76	24.40	2.93	12.81			
		7.58	17.92	2.93	0.00	0.0581		
				2.98	13.30			
				2.98	0.00	0.0793		
				3.03	13.79			
				3.03	0.00	0.0693		
				3.08	14.30			
				3.08	0.00	0.0661		
				3.13	14.81			
				3.13	0.00	0.0645		
				3.18	15.34			
				3.18	0.00	0.0645		
				3.23	15.87			
				3.23	0.00	0.0672		
				3.28	16.42			
				3.28	0.00	0.0700		
				3.33	16.97			
				3.33	0.00	0.0817		
				3.38	17.53			
				3.38	0.00	0.0729		
				3.43	18.10			
				3.43	0.00	0.0699		
				3.48	18.68			
				3.48	0.00	0.0731		

APPENDIX 3. Water flow data and breakthrough turbidity data

Lysimeter A continued								
Time	Total outflow			Time	Accum. flow	Out/In (turb.)		
(h in dec.fc	[mm/h]			(h in dec.fc	[mm]			
				3.53	19.26			
				3.53	0.00	0.0708		
				3.58	19.85			
				3.58	0.00	0.0709		
				3.63	20.45			
				3.63	0.00	0.0736		
				3.68	21.05			
				3.68	0.00	0.0920		
				3.73	21.66			
				3.73	0.00	0.0761		
				3.78	22.28			
				3.78	0.00	0.0761		
				3.83	22.91			
				3.83	0.00	0.0784		
				3.88	23.55			
				3.88	0.00	0.0777		
				3.93	24.20			
				3.93	0.00	0.0769		
				3.98	24.86			
				3.98	0.00	0.0799		
				4.03	25.53			
				4.03	0.00	0.0857		
				4.08	26.20			
				4.08	0.00	0.0860		
				4.13	26.89			
				4.13	0.00	0.0893		
				4.18	27.58			
				4.18	0.00	0.1051		
				4.23	28.28			
				4.23	0.00	0.0897		
				4.28	28.98			
				4.28	0.00	0.0873		
				4.33	29.70			
				4.33	0.00	0.0912		
				4.40	30.42			
				4.43	0.00	0.0828		
				4.48	31.14			
				4.48	0.00	0.0644		
				4.58	31.83			
				4.58	0.00	0.0824		
				4.71	32.42			
				4.73	0.00	0.0688		
				4.91	32.80			
				4.91	0.00	0.0566		
				5.11	33.01			
				5.11	0.00	0.0402		
				5.28	33.12			
				5.28	0.00	0.0297		
				5.53	33.17			
				5.53	0.00	0.0259		
				5.88	33.17			
				5.88	0.00	0.0210		

APPENDIX 3. Water flow data and breakthrou[illegible] turbidity data

Lysimeter A	continued		
Time	Total outflow		
(h in dec.fc	[mm/h]		

T[illegible]e	Accum. flow	Out/In (turb.)
(h in[illegible]c.fc	[mm]	
0.26	33.17	
0.26	0.00	0.0132
0.73	33.17	
0.73		0.0130

APPENDIX 3. Water flow data and breakthrough turbidity data

Lysimeter B								
Time	Tot.outflow	Accum.flow	Time	Turbidity	Time	Out/In (turbidity)		
(h in dec.fo	[mm/h]	[mm]	in dec.forn	[NTU]	(h in dec.form)			
2.22	4.10	0.15	1.92	208.75	1.916667	0.114887		
2.32	4.64	0.59	2.22	51.57	0	0		
2.42	4.64	1.05	2.32	11.33	0	0		
2.47	5.44	1.30	2.42	11.25	0	0		
2.53	5.44	1.67	2.47	11.57	2.216667	0.02838		
2.60	5.44	2.03	2.53	12.20	2.316667	0.006237		
2.67	5.44	2.39	2.60	14.30	2.416667	0.006192		
2.73	5.44	2.76	2.67	14.45	0	0		
2.80	5.44	3.12	2.73	15.33	2.466667	0.006366		
2.87	5.57	3.49	2.80	15.47	2.533333	0.006714		
2.93	5.57	3.86	2.87	16.55	2.6	0.00787		
3.00	5.75	4.23	2.93	16.83	2.666667	0.007953		
3.07	5.93	4.62	3.00	17.77	2.733333	0.008439		
3.13	5.62	5.01	3.07	18.07	2.8	0.008512		
3.20	5.66	5.38	3.13	18.80	2.866667	0.009108		
3.27	5.68	5.76	3.20	19.17	2.933333	0.00926		
3.33	5.71	6.14	3.27	19.67	3	0.009778		
3.40	5.77	6.52	3.33	19.73	3.066667	0.009943		
3.47	5.55	6.90	3.40	20.93	3.133333	0.010347		
3.53	5.82	7.28	3.47	22.00	3.2	0.010549		
3.60	5.51	7.66	3.53	23.17	3.266667	0.010824		
3.67	5.53	8.03	3.60	24.00	3.333333	0.01086		
3.73	5.62	8.40	3.67	26.33	3.4	0.011516		
3.80	5.62	8.77	3.73	28.53	3.466667	0.012108		
3.87	5.62	9.15	3.80	29.70	3.533333	0.01275		
3.93	5.62	9.52	3.87	29.07	3.6	0.013209		
4.00	5.53	9.89	3.93	29.67	3.666667	0.014493		
4.07	5.53	10.26	4.00	29.60	3.733333	0.015704		
4.13	5.64	10.63	4.13	30.74	3.8	0.016346		
4.20	5.49	11.01	4.27	30.23	3.866667	0.015997		
4.27	5.49	11.37	4.33	33.77	3.933333	0.016327		
4.33	5.33	11.73	4.40	33.67	4	0.016291		
4.40	5.33	12.09	4.53	32.93	0	0		
4.47	5.31	12.44	4.67	32.68	4.133333	0.016918		
4.53	5.18	12.79	4.73	35.70	0	0		
4.60	4.92	13.13	4.87	33.57	4.266667	0.016635		
4.67	4.92	13.46	5.00	34.80	4.333333	0.018584		
4.73	5.07	13.79	5.20	36.78	4.4	0.018529		
4.80	5.07	14.13	5.40	42.38	0	0		
4.87	4.87	14.46	5.54	57.73	4.533333	0.018125		
4.93	4.83	14.78	5.71	56.50	0	0		
5.00	4.81	15.10	5.79	59.30	4.666667	0.017983		
5.07	4.70	15.42	5.88	65.16	4.733333	0.019648		
5.13	4.67	15.73	6.04	79.90	0	0		
5.20	4.67	16.04	6.13	83.40	4.866667	0.018474		
5.27	4.61	16.35	6.29	88.83	0	0		
5.33	4.85	16.67	6.46	102.33	5	0.019152		
5.40	4.85	16.99	6.54	103.75	0	0		
5.48	4.76	17.35	6.71	116.67	0	0		
5.56	4.88	17.75	6.88	124.86	5.2	0.020239		
5.64	4.88	18.16	6.96	127.33	0	0		
5.73	4.88	18.57	7.04	127.40	0	0		

APPENDIX 3. Water flow data and breakthrough turbidity data

Lysimeter B		continued						
Time	Total outflow	Accum.flow	Time	Turbidity	Time	Out/In (turbidity)		
(h in dec.fc	[mm/h]	[mm]	(h in dec.fc	[NTU]	(h in dec.fc	0.031769		
5.81	4.85	18.97	7.13	144.00	5.4	0.023321		
5.89	4.93	19.38	7.21	156.00	0	0		
5.98	5.06	19.80	7.29	164.00	5.541667	0.031769		
6.06	5.02	20.22	7.38	171.67	0	0		
6.14	5.02	20.63	7.46	172.80	5.708333	0.031095		
6.23	5.02	21.05	7.54	171.75	5.791667	0.032636		
6.31	5.02	21.47	7.63	171.75	5.875	0.035861		
6.39	5.02	21.89	7.71	179.75	0	0		
6.48	5.02	22.31	7.79	201.75	6.041667	0.043974		
6.56	5.02	22.73	7.88	210.67	6.125	0.0459		
6.64	5.11	23.15	7.96	226.25	0	0		
6.73	5.11	23.57	8.04	231.40	6.291667	0.048886		
6.81	5.11	24.00	8.13	245.00	0	0		
6.89	5.11	24.43	8.21	245.20	6.458333	0.05632		
6.98	5.20	24.86	8.29	226.50	6.541667	0.0571		
7.06	5.20	25.29	8.38	222.67	0	0		
7.14	5.28	25.73	8.46	203.00	6.708333	0.064208		
7.23	5.28	26.17	8.54	206.00	0	0		
7.31	5.28	26.61	8.63	239.00	6.875	0.068716		
7.39	5.30	27.05	8.71	245.00	6.958333	0.070079		
7.48	5.30	27.49	8.79	251.13	7.041667	0.070116		
7.56	5.30	27.93	8.88	249.83	7.125	0.079252		
7.64	5.30	28.37	8.96	255.00	7.208333	0.085856		
7.73	5.43	28.82	9.04	249.00	7.291667	0.090259		
7.81	5.43	29.27	9.13	252.60	7.375	0.094478		
7.89	5.51	29.73	9.21	251.50	7.458333	0.095102		
7.98	5.51	30.19	9.29	255.86	7.541667	0.094524		
8.06	5.44	30.64	9.38	258.75	7.625	0.094524		
8.14	5.44	31.10	9.46	281.50	7.708333	0.098927		
8.23	5.44	31.55	9.54	263.33	7.791667	0.111035		
8.31	5.44	32.00	9.62	284.17	7.875	0.115942		
8.39	5.44	32.46	9.68	240.83	7.958333	0.124518		
8.48	5.44	32.91	9.75	220.33	8.041667	0.127353		
8.56	5.41	33.36	9.82	207.00	8.125	0.134838		
8.64	5.41	33.81	9.88	181.00	8.208333	0.134948		
8.73	5.48	34.27	9.95	153.75	8.291667	0.124656		
8.81	5.48	34.72	10.08	120.50	8.375	0.122546		
8.89	5.41	35.18	10.22	98.18	8.458333	0.111723		
8.98	5.41	35.63	10.35	80.84	8.541667	0.113374		
9.06	5.34	36.08	10.55	70.34	8.625	0.131535		
9.14	5.34	36.52	10.75	57.60	8.708333	0.134838		
9.23	5.46	36.97	11.02	42.60	8.791667	0.138209		
9.31	5.46	37.43	11.15	41.90	8.875	0.137498		
9.39	5.34	37.88	11.55	18.15	8.958333	0.140341		
9.48	5.34	38.32	12.48	9.87	9.041667	0.137039		
9.56	5.14	38.76	13.22	11.25	9.125	0.13902		
9.63	3.81	39.09			9.208333	0.138415		
9.70	4.37	39.37			9.291667	0.140813		
9.77	4.04	39.65			9.375	0.142405		
9.83	3.86	39.91			9.458333	0.154926		
9.90	3.51	40.15			9.541667	0.144928		
9.97	3.23	40.38			9.616667	0.156393		

APPENDIX 3. Water flow data and breakthrough turbidity data

Lysimeter B		continued						
Time	Total outflow	Accum.flow			Time	Out/In (turbidity)		
(h in dec.fc	[mm/h]	[mm]			(h in dec.fc	0		
10.03	3.01	40.59			9.683333	0.132544		
10.10	2.79	40.78			9.75	0.121262		
10.17	2.61	40.96			9.816667	0.113924		
10.23	2.61	41.13			9.883333	0.099615		
10.30	2.61	41.31			9.95	0.084618		
10.37	2.13	41.47			0	0		
10.43	1.93	41.60			10.08333	0.066318		
10.50	1.82	41.73			0	0		
10.57	1.65	41.84			10.21667	0.054034		
10.63	1.65	41.95			10.35	0.044491		
10.70	1.14	42.05			10.55	0.038712		
10.77	1.03	42.12			10.75	0.031701		
10.83	0.88	42.18			11.01667	0.023445		
10.90	0.88	42.24			11.15	0.02306		
10.97	0.88	42.30			11.55	0.009989		
11.03	0.81	42.35			12.48333	0.005432		
11.10	2.59	42.47			13.21667	0.006192		
11.17	2.06	42.62						
11.23	0.07	42.69						
11.30	0.35	42.71						
11.37	0.44	42.73						
11.43	0.44	42.76						
11.50	0.31	42.79						
11.57	0.31	42.81						
11.63	0.29	42.83						
11.70	0.29	42.85						
11.77	0.20	42.86						
11.83	0.20	42.88						
11.90	0.20	42.89						
11.97	0.20	42.90						
12.03	0.20	42.92						
12.10	0.20	42.93						
12.17	0.18	42.94						
12.23	0.18	42.95						
12.30	0.18	42.97						
12.37	0.18	42.98						
12.43	0.18	42.99						
12.50	0.18	43.00						
12.57	0.18	43.01						
12.63	0.18	43.02						
12.70	0.18	43.04						
12.77	0.18	43.05						
12.83	0.18	43.06						
12.90	0.18	43.07						
12.97	0.07	43.08						
13.03	0.07	43.08						
13.10	0.07	43.09						
13.17	0.07	43.09						
13.23	0.07	43.10						

APPENDIX 3. Water flow data and breakthrough turbidity data

Lysimeter C						
Time	Total outflow	Accum.flow	Time	Turbidity	Time	Out/In (turbidity)
(h in dec.fo	[mm/h]	[mm]	h in dec.form	[NTU]	(h in dec.form)	
1.28	3.00	0.06	1.03	164.00	1.03	0.092
1.47	6.14	0.90	1.28	90.44	1.28	0.051
1.55	13.21	1.71	1.36	43.05	1.36	0.024
1.78	14.18	4.90	1.43	51.50	1.43	0.029
1.92	13.71	6.76	1.50	75.55	1.50	0.042
2.08	11.87	8.89	1.57	43.60	1.57	0.024
2.21	12.25	10.40	1.63	28.20	1.63	0.016
2.36	12.65	12.27	1.70	26.70	1.70	0.015
2.66	12.96	16.11	1.77	14.85	1.77	0.008
2.96	13.39	20.06	1.90	15.40	1.90	0.009
3.08	13.49	21.74	2.10	15.90	2.10	0.009
3.21	13.43	23.43	2.23	18.35	2.23	0.010
3.33	13.47	25.00	2.43	22.30	2.43	0.013
3.63	13.21	29.00	2.63	51.05	2.63	0.029
3.93	13.14	32.95	2.83	76.70	2.83	0.043
4.07	13.34	34.83	2.97	86.60	2.97	0.049
4.21	13.52	36.73	3.10	113.50	3.10	0.064
4.43	13.80	39.80	3.30	132.00	3.30	0.074
4.56	13.88	41.53	3.57	166.50	3.57	0.093
4.74	13.88	44.08	3.77	177.00	3.77	0.099
4.89	13.76	46.15	3.90	195.00	3.90	0.109
4.99	13.34	47.50	4.03	213.00	4.03	0.119
5.09	13.21	48.83	4.17	224.50	4.17	0.126
5.19	12.97	50.14	4.30	243.50	4.30	0.137
5.31	13.29	51.67	4.50	277.50	4.50	0.156
5.42	13.78	53.25	4.70	297.50	4.70	0.167
5.63	13.92	56.14	4.83	303.50	4.83	0.170
5.83	13.34	58.86	4.97	327.50	4.97	0.184
5.88	13.39	59.42	5.17	345.00	5.17	0.193
5.95	13.56	60.43	5.30	340.00	5.30	0.191
6.05	13.52	61.79	5.43	358.50	5.43	0.201
6.15	13.67	63.15	5.57	371.00	5.57	0.208
6.25	13.50	64.50	5.70	388.50	5.70	0.218
6.32	13.08	65.39	5.83	404.00	5.83	0.227
6.38	13.32	66.27	5.97	414.50	5.97	0.232
6.45	12.73	67.14	6.10	416.00	6.10	0.233
6.52	11.41	67.94	6.23	422.50	6.23	0.237
6.58	8.62	68.61	6.37	409.50	6.37	0.230
6.65	5.20	69.07	6.50	371.00	6.50	0.208
6.72	3.51	69.36	6.63	318.50	6.63	0.179
6.88	1.78	69.80	6.70	471.00	6.70	0.264
7.10	0.99	70.10	6.77	265.50	6.77	0.149
7.43	1.18	70.46	6.93	188.00	6.93	0.105
7.78	0.83	70.82	7.10	116.00	7.10	0.065
7.78	1.09	70.82	7.30	64.40	7.30	0.036
8.08	0.41	71.04	7.47	44.55	7.47	0.025
8.68	0.39	71.28	7.63	45.60	7.63	0.026
9.58	0.29	71.59	8.07	20.45	8.07	0.011
10.08	0.26	71.72	8.60	11.32	8.60	0.006
10.22	0.26	71.76				
10.35	0.26	71.79				
11.22	0.24	72.01				

APPENDIX 3. Water flow data and breakthrough turbidity data

Lysimeter C		continued					
Time	Total outflow	Accum.flow					
(h in dec.fc	[mm/h]	[mm]					
11.45	0.18	72.06					
11.65	0.11	72.09					
11.95	0.13	72.13					
12.92	0.13	72.25					
13.12	0.13	72.28					

APPENDIX 3. Water flow data and breakthrough turbidity data

Lysimeter D							
Time	Total outflow	Accum.flow	Time	Turbidity	Time	Out/In (turbidity)	
(h in dec.fo	[mm/h]	[mm]	(h in dec.fo	[NTU]	(h in dec.form)		
1.66	6.27	0.17	1.66	16.38	1.66	0.009	
1.74	11.89	0.93	1.71	14.15	1.71	0.008	
1.78	10.41	1.36	1.76	14.85	1.76	0.008	
1.85	9.79	2.04	1.86	18.33	1.86	0.010	
1.91	10.58	2.66	1.91	12.67	1.91	0.007	
1.98	10.21	3.38	1.96	9.84	1.96	0.006	
2.07	5.72	4.11	2.01	8.91	2.01	0.005	
2.16	11.82	4.94	2.11	13.45	2.11	0.008	
2.25	8.17	5.80	2.16	11.20	2.16	0.006	
2.34	9.00	6.61	2.21	10.80	2.21	0.006	
2.43	8.46	7.39	2.36	12.37	2.36	0.007	
2.53	5.60	8.03	2.41	12.70	2.41	0.007	
2.61	10.87	8.69	2.46	11.07	2.46	0.006	
2.68	7.94	9.43	2.56	13.83	2.56	0.008	
2.76	8.26	10.08	2.61	10.63	2.61	0.006	
2.84	10.83	10.85	2.66	9.95	2.66	0.006	
2.93	5.44	11.57	2.76	15.60	2.76	0.009	
3.02	11.65	12.33	2.81	10.90	2.81	0.006	
3.12	6.32	13.18	2.86	9.75	2.86	0.006	
3.20	10.48	13.85	2.91	12.47	2.91	0.007	
3.29	7.70	14.69	3.01	11.50	3.01	0.006	
3.41	6.69	15.55	3.06	11.13	3.06	0.006	
3.52	5.77	16.22	3.16	14.70	3.16	0.008	
3.64	9.64	17.21	3.21	12.13	3.21	0.007	
3.74	6.42	17.99	3.26	11.10	3.26	0.006	
3.84	11.12	18.84	3.31	13.25	3.31	0.007	
3.93	7.65	19.73	3.41	13.00	3.41	0.007	
4.04	8.17	20.61	3.46	12.97	3.46	0.007	
4.16	6.97	21.47	3.56	15.37	3.56	0.009	
4.26	6.12	22.10	3.61	13.73	3.61	0.008	
4.35	8.39	22.79	3.66	13.00	3.66	0.007	
4.44	6.22	23.48	3.81	15.70	3.81	0.009	
4.53	9.77	24.19	3.86	15.00	3.86	0.008	
4.64	8.74	25.19	3.96	19.03	3.96	0.011	
4.75	7.43	26.07	4.01	18.60	4.01	0.010	
4.86	8.47	26.91	4.11	22.90	4.11	0.013	
4.96	2.98	27.50	4.16	20.53	4.16	0.012	
5.06	9.37	28.15	4.21	21.90	4.21	0.012	
5.17	3.84	28.86	4.31	22.83	4.31	0.013	
5.28	9.22	29.55	4.36	21.77	4.36	0.012	
5.39	5.62	30.36	4.41	23.75	4.41	0.013	
5.50	7.98	31.15	4.46	31.03	4.46	0.018	
5.61	6.78	31.91	4.51	25.75	4.51	0.015	
5.70	5.74	32.49	4.56	22.88	4.56	0.013	
5.79	8.20	33.14	4.61	23.16	4.61	0.013	
5.88	5.97	33.79	4.66	25.85	4.66	0.015	
5.97	9.62	34.49	4.76	27.04	4.76	0.015	
6.08	4.77	35.23	4.81	24.94	4.81	0.014	
6.15	10.39	35.81	4.86	23.97	4.86	0.014	
6.24	8.08	36.64	4.96	32.77	4.96	0.018	
6.35	6.10	37.40	5.01	26.43	5.01	0.015	
6.44	8.17	38.08	5.06	25.13	5.06	0.014	

APPENDIX 3. Water flow data and breakthrough turbidity data

Lysimeter D		continued				
Time	Total outflow	Accumulated	Time	Turbidity	Time	Out/In (turbidity)
(h in dec.fc	[mm/h]	[mm]	(h in dec.fc	[NTU]	in dec.form)	
6.51	4.01	38.50	5.11	25.58	5.11	0.014
6.59	9.73	39.05	5.21	31.25	5.21	0.018
6.68	7.17	39.81	5.26	28.63	5.26	0.016
6.77	7.72	40.47	5.31	27.83	5.31	0.016
6.86	8.78	41.20	5.36	28.00	5.36	0.016
6.96	5.40	41.89	5.46	39.57	5.46	0.022
7.06	6.45	42.50	5.51	30.04	5.51	0.017
7.18	9.25	43.44	5.56	31.18	5.56	0.018
7.28	4.14	44.13	5.71	43.85	5.71	0.025
7.34	9.60	44.54	5.76	39.25	5.76	0.022
7.40	8.61	45.08	5.91	51.84	5.91	0.029
7.47	6.51	45.56	5.96	46.98	5.96	0.027
7.54	7.85	46.08	6.01	43.93	6.01	0.025
7.63	9.02	46.85	6.11	59.48	6.11	0.034
7.73	8.84	47.77	6.16	48.90	6.16	0.028
7.83	3.96	48.39	6.21	45.70	6.21	0.026
7.91	10.63	48.96	6.26	46.77	6.36	0.036
8.01	6.46	49.81	6.36	63.70	6.41	0.031
8.11	7.82	50.53	6.41	55.70	6.46	0.030
8.22	10.28	51.56	6.46	53.33	6.61	0.041
8.31	8.25	52.36	6.61	73.16	6.66	0.037
8.43	6.70	53.23	6.66	66.30	6.81	0.044
8.56	6.14	54.08	6.81	77.23	6.86	0.040
8.67	7.56	54.83	6.86	70.50	7.01	0.044
8.79	6.30	55.67	7.01	78.70	7.06	0.045
8.89	10.29	56.53	7.06	79.93	7.16	0.046
9.00	6.36	57.40	7.16	81.45	7.21	0.048
9.11	5.48	58.06	7.21	84.38	7.26	0.046
9.26	6.03	58.92	7.26	81.83	7.36	0.056
9.37	8.99	59.78	7.36	99.55	7.41	0.051
9.51	5.68	60.78	7.41	90.00	7.46	0.049
9.66	4.88	61.57	7.46	85.95	7.56	0.053
9.78	9.06	62.44	7.56	94.20	7.61	0.051
9.91	5.31	63.34	7.61	91.20	7.71	0.067
10.04	6.54	64.13	7.71	118.00	7.76	0.060
10.16	6.86	64.91	7.76	106.33	7.81	0.058
10.31	6.26	65.90	7.81	102.67	7.91	0.067
10.46	6.30	66.84	7.91	118.50	7.96	0.067
10.57	6.13	67.51	7.96	118.75	8.06	0.073
10.71	6.06	68.38	8.06	130.00	8.11	0.071
10.86	5.98	69.28	8.11	125.50	8.21	0.084
10.97	10.76	70.19	8.21	148.80	8.26	0.078
11.04	7.68	70.90	8.26	137.60	8.41	0.082
11.14	8.65	71.65	8.41	145.50	8.56	0.088
11.25	7.91	72.62	8.56	156.50	8.61	0.088
11.36	6.11	73.36	8.61	156.75	8.71	0.095
11.46	9.36	74.13	8.71	169.00	8.76	0.092
11.57	8.27	75.09	8.76	163.50	8.86	0.101
11.70	7.09	76.13	8.86	178.75	9.01	0.109
11.81	5.94	76.82	8.91	173.67	9.06	0.100
11.91	9.49	77.61	9.01	192.33	9.21	0.109
12.02	8.61	78.59	9.06	177.25	9.26	0.106

APPENDIX 3. Water flow data and breakthrough turbidity data

Lysimeter D		continued					
Time	Total outflow	Accumulated	Time	Turbidity	Time	Out/In (turbidity)	
(h in dec.fo	[mm/h]	[mm]	(h in dec.fo	[NTU]	in dec.form)		
12.16	5.20	79.55	9.21	192.50	9.36	0.114	
12.31	7.72	80.50	9.26	187.33	9.41	0.110	
12.41	4.49	81.13	9.36	202.33	9.51	0.120	
12.56	5.34	81.87	9.41	195.75	9.56	0.108	
12.69	9.06	82.79	9.51	212.25	9.61	0.102	
12.81	4.68	83.63	9.56	192.00	9.76	0.105	
12.91	10.13	84.37	9.61	181.50	9.81	0.096	
13.04	8.18	85.56	9.76	185.50	9.96	0.112	
13.16	5.17	86.36	9.81	169.75	10.01	0.108	
13.28	8.40	87.15	9.96	199.00	10.11	0.130	
13.40	5.90	88.06	10.01	191.00	10.16	0.125	
13.51	5.53	88.67	10.11	230.00	10.21	0.125	
13.62	8.58	89.45	10.16	221.25	10.31	0.132	
13.71	5.42	90.09	10.21	221.33	10.36	0.133	
13.79	9.35	90.69	10.31	234.50	10.46	0.141	
13.88	3.32	91.22	10.36	236.00	10.51	0.136	
14.01	6.48	91.87	10.46	250.00	10.61	0.155	
14.16	1.36	92.46	10.51	240.50	10.66	0.141	
14.31	2.36	92.74	10.61	274.80	10.76	0.165	
14.46	1.94	93.06	10.66	250.00	10.81	0.151	
14.61	2.16	93.37	10.76	292.25	10.96	0.164	
14.76	0.92	93.60	10.81	266.75	11.11	0.169	
14.91	1.06	93.75	10.96	290.20	11.16	0.168	
15.06	2.50	94.01	11.11	300.00	11.26	0.171	
15.21	0.79	94.26	11.16	298.00	11.31	0.170	
15.36	0.63	94.37	11.26	302.67	11.41	0.181	
15.51	0.67	94.46	11.31	301.25	11.46	0.173	
15.66	2.29	94.69	11.41	321.25	11.56	0.181	
15.81	1.23	94.95	11.46	307.25	11.61	0.175	
15.96	1.14	95.13	11.56	320.50	11.71	0.185	
16.11	1.05	95.29	11.61	310.25	11.76	0.181	
16.26	3.00	95.60	11.71	328.25	11.86	0.194	
16.41	2.18	95.98	11.76	321.00	11.91	0.185	
16.56	0.87	96.21	11.86	343.00	12.01	0.198	
16.71	0.74	96.33	11.91	327.00	12.06	0.204	
16.86	0.81	96.45	12.01	350.50	12.11	0.220	
17.01	0.50	96.55	12.06	361.00	12.16	0.205	
17.16	0.48	96.62	12.11	389.25	12.26	0.209	
17.34	0.45	96.71	12.16	362.50	12.31	0.205	
17.46	0.41	96.76	12.26	371.00	12.41	0.204	
17.55	0.40	96.80	12.31	364.00	12.51	0.211	
			12.41	361.75	12.56	0.197	
			12.51	373.33	12.66	0.204	
			12.56	348.75	12.71	0.196	
			12.66	361.67	12.81	0.204	
			12.71	348.00	12.91	0.217	
			12.81	361.33	12.96	0.216	
			12.91	383.75	13.06	0.218	
			12.96	382.67	13.16	0.236	
			13.06	386.67	13.21	0.226	
			13.16	418.50	13.26	0.239	
			13.21	400.67	13.31	0.229	

APPENDIX 3. Water flow data and breakthrough turbidity data

Lysimeter D		continued					
Time	Total outflow	Accumulated	Time	Turbidity	Time	Out/In (turbidity)	
(h in dec.fo	[mm/h]	[mm]	(h in dec.fo	[NTU]	in dec.form)		
			13.26	424.00	13.41	0.237	
			13.31	406.25	13.56	0.238	
			13.41	420.00	13.66	0.241	
			13.56	421.67	13.81	0.239	
			13.66	426.60	13.96	0.229	
			13.81	422.67	14.01	0.211	
			13.96	405.50	14.03	0.212	
			14.01	373.20	14.26	0.203	
			14.03	375.50	14.28	0.171	
			14.26	359.80	14.41	0.127	
			14.28	302.75	14.56	0.102	
			14.41	224.50	14.71	0.084	
			14.56	180.25	14.83	0.085	
			14.71	148.25	15.26	0.033	
			14.83	150.67	15.41	0.026	
			15.26	57.60	15.96	0.010	
			15.41	45.87	16.21	0.006	
			15.96	17.90	16.56	0.004	
			16.21	10.20	17.01	0.003	
			16.56	7.53			
			17.01	5.80			

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